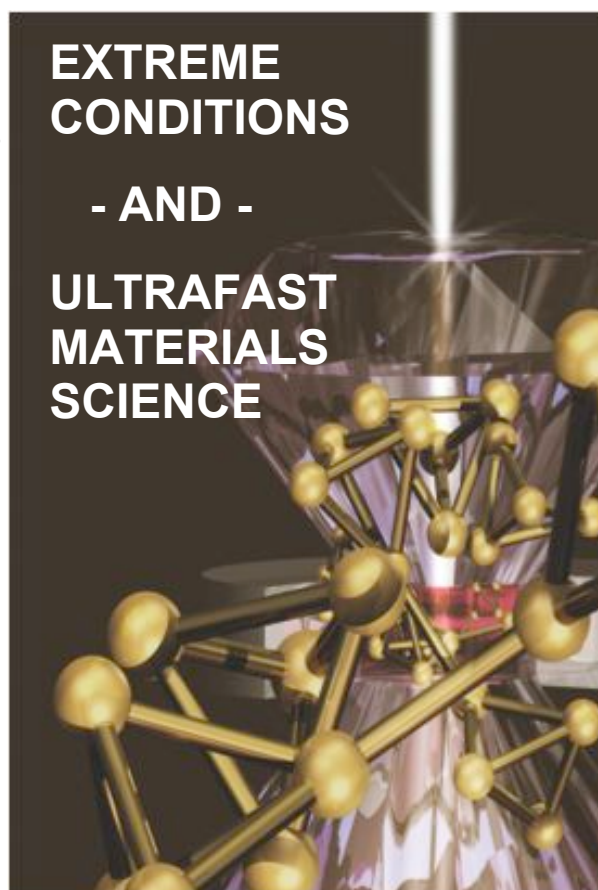


EXTREME CHEMISTRY GROUP

Our experimental research group is composed of four Ph.D.s with expertise in condensed matter physics, physical chemistry, and ultrafast laser physics phenomena. We have in excess of 50 years professional experience with 100+ publications in predominately top-flight peer reviewed journals. We also design, build, and test laser-based (non-contact) tabletop diagnostic tools to address *Grand Challenge* scientific issues.



Mike Armstrong



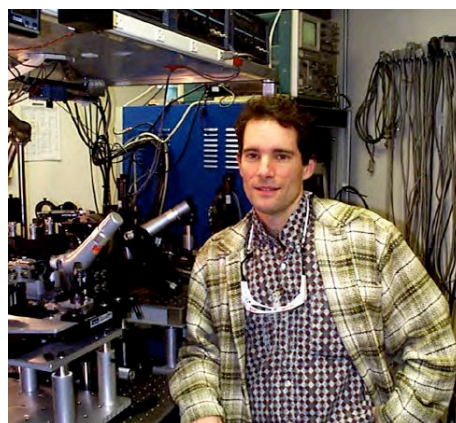
Jonathan Crowhurst



Chris Grant



Joe Zaug



This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

LLNL-Physical & Life Sciences, Chemical Sciences Division, Extreme Chemistry Group

Mike Armstrong, Ph.D.

armstrong30@llnl.gov, 925-423-5702

Jonathan Crowhurst, Ph.D.

crowhurst1@llnl.gov, 925-422-1945

Christian Grant, Ph.D.

grant29@llnl.gov, 925-424-5951

Joseph M. Zaug (Group Founder), Ph.D.

zaug1@llnl.gov, 925-423-4428

Our Experimental Capabilities and Expertise: An Overview

Sample conditions we can achieve are,

Pressure, 0-100+ GPa using custom-designed diamond-anvil cells (DACs)
Temperature, 300 K – 4000+ K, resistive heating, internal heating, laser heating
Temperatures, 1+ eV delivered in 100s of nanoseconds, capacitor driven arc

Experimental diagnostics and techniques: designed, built, and tested in-house at LLNL

- Speed of sound, laser-induced ultrasonics (0.5 – 10 GHz range), ps ISLS, PALS
- Thermal transport, Impulsive Stimulated Light Scattering, ps ISLS
- Chemical and Mass transport, ns ISLS
- Electronic and structural relaxation rates, ns, ps ISLS
- Electrical transport, four-probe DAC (Collaboration with S. T. Weir)
- ns pulse-gated Raman (420 - 840 nm pump range, 1-5 kHz rate, 20 ns pulse width)
- Vibrational spectroscopy, IR (500 – 10,000 cm^{-1}), Raman (457 – 840 nm pump)
- Fluorescence spectroscopy (457 – 840 nm pump)
- UV-VIS transmission, (225 – 950 nm range)
- X-ray diffraction/scattering (10-80 keV) + simultaneous laser heating to 3500 K*
- Neutron scattering/diffraction at LANL-LANSCE*
- Laser-induced reaction propagation rates, with Streak camera (10 – 3000 m/sec)
- High P-T material synthesis in a DAC
- High P-T SERS, enhancement for Raman spectroscopy
- fs-ns laser shock induced studies, single-shot, ultrafast shock interrogation, USI
- fs laser shock initiation; shock propagation or speed of sound measurements, USI
- fs pulsed TRIR (50 fs – 1ms time resolution range, single-shot)

Ambient condition diagnostics on recovered samples

- Nearly all known analytical capabilities are available at LLNL and SNL

Potential future experimental developments

- A myriad of fs pump-probe techniques e.g., aq. e^- solvation, photo-pumped phase transitions and disordering, phonon relaxation rates.

Advanced designs completed for future material studies

VUV transmission (150 – 350 nm), tabletop instrument for high P-T material studies

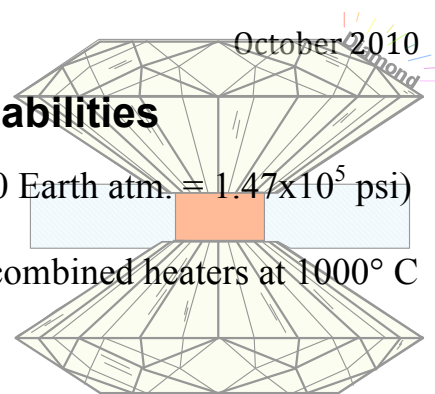
* Off site synchrotron and spallation reactor capabilities in U.S. and abroad.

Reaching Extreme Conditions: High P-T Capabilities

Pressure Range: 0.1 - 100+ GPa (1 GPa=10 kbar = 10,000 Earth atm. $\approx 1.47 \times 10^5$ psi)

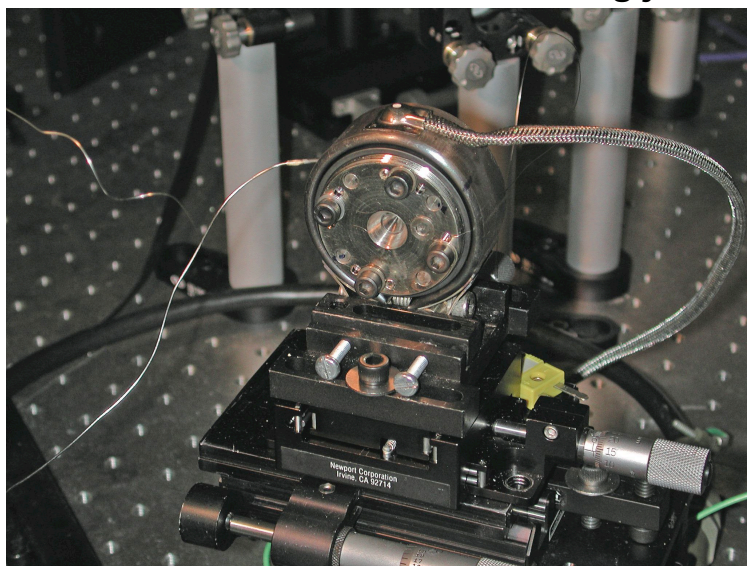
Temperature Range:

- External Resistive Heating, 1 focused-heater at 600° C; 2 combined heaters at 1000° C
- Internal Resistive Heating, up to 3500 K (3773°C)
- Laser Heating, > 3000 K (>3273°C); Spark heating > 1 eV

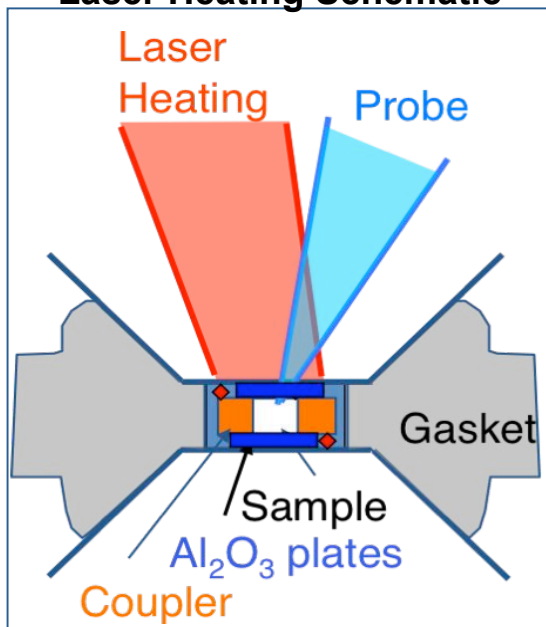


We developed high-strength at temperature diamond-anvil cells (DACs) made from the nickel-based superalloys Inconel 718 and Nimonic 80A. These fully hardened platens may be safely heated to 973 K, and 1083 (700° C, 810° C). Our resistive focused-heater achieves a sample temperature of 600 °C where the DAC platens remain < 300 °C. Our internal resistive heater is embedded within a high-pressure sample to achieved 3500 K at pressures up to 30 GPa. Our 60 Watt Nd:YAG fiber laser heating system can achieve > 3000 K at > 50 GPa sample pressure. Arc heating achieves 1+ eV temperatures in 100s of nanoseconds.

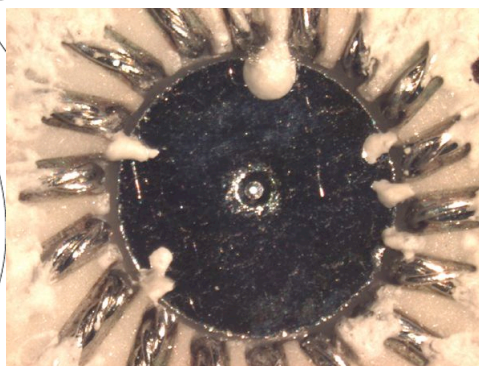
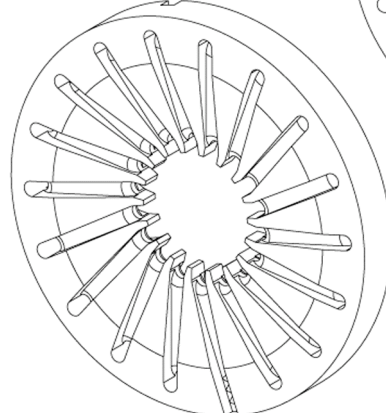
Inconel 718 DAC & external heating jacket



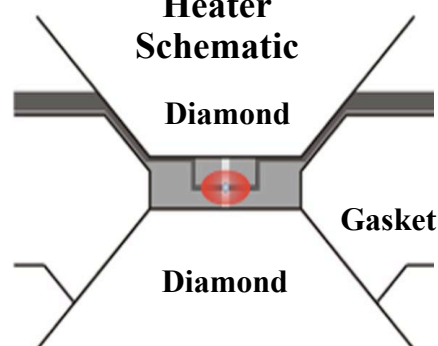
Laser Heating Schematic



The resistive focused-heater surrounds the sample chamber



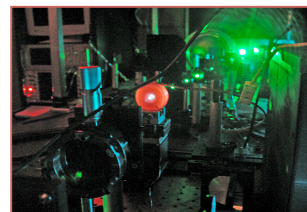
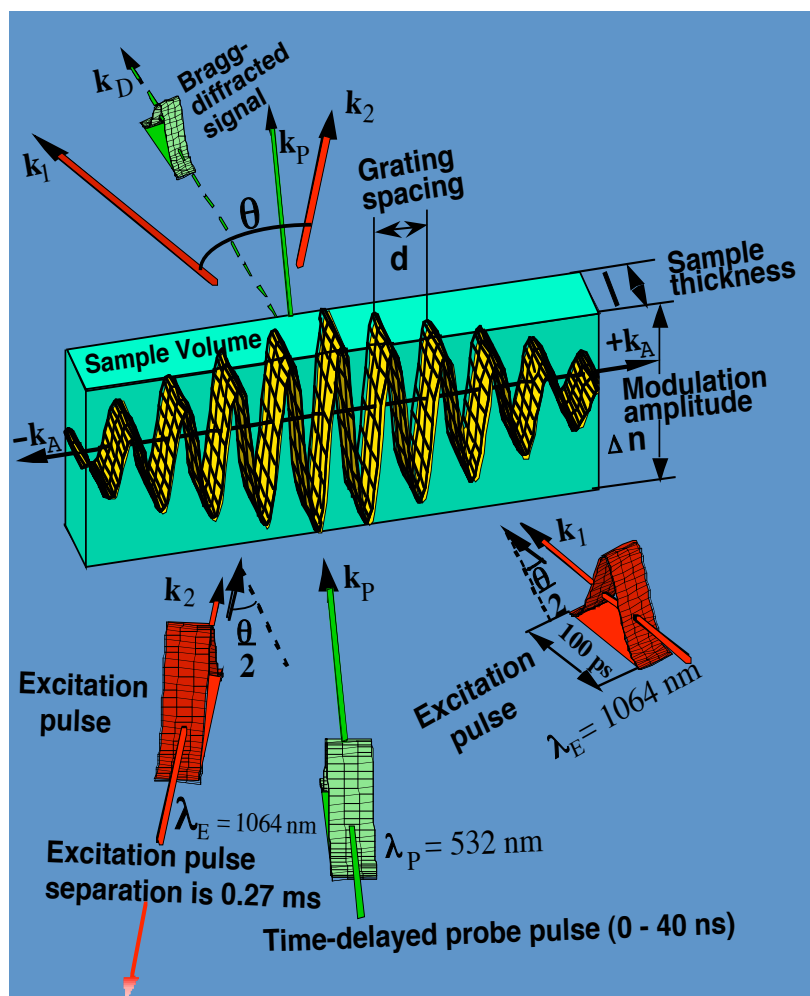
Internal Heater Schematic



Information Contacts: Joe Zaug (zaug1@llnl.gov) or Jonathan Crowhurst (Crowhurst1@llnl.gov)

Impulsive Stimulated Light Scattering (ISLS)

Speed of Sound; 1D Thermal Diffusivity


 ber 2010
**ISLS of
 H₂O at 6
 GPa and
 800 K**


ISLS is performed to generate and measures acoustic wave propagation within a wide class of transparent and opaque materials. 1-D thermal transport is measured from transparent materials, though in principal, opaque materials are amenable to the ISLS technique. A preselected acoustic grating ($d=1-10\ \mu\text{m}$) is measured using a calibrated isotropic standard. Fourier transformation of measured time-domain spectra yield frequency values with better than 1% precision. The acoustic frequency range of ISLS is limited only by the excitation laser pulse-width, e.g. a 100 ps pulse provides a $<10\ \text{GHz}$ range. A custom-built μ -Raman spectroscopy system is embedded within our ISLS instrument to facilitate near-

simultaneous structural and/or chemical phase information.

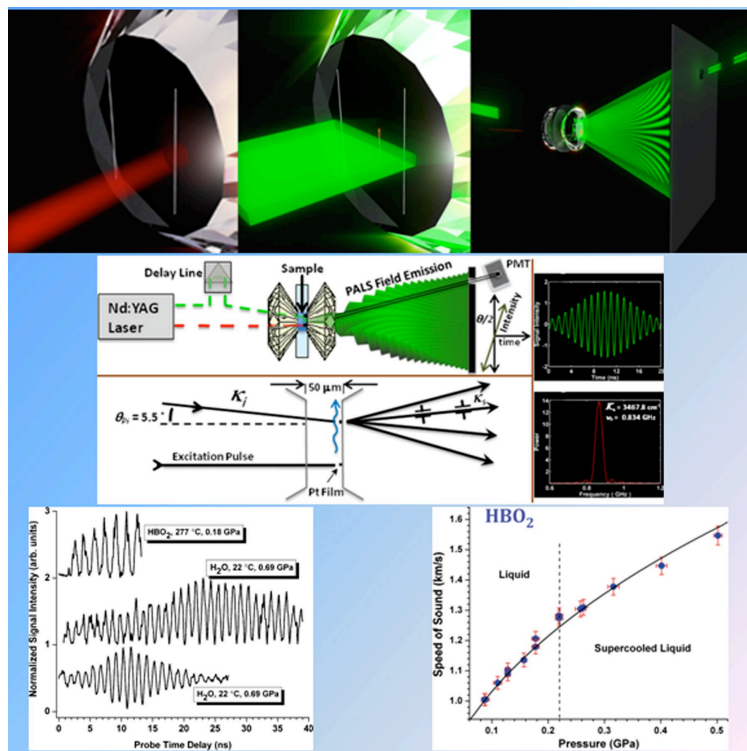
Selected ISLS publications:

1. J. C. Crowhurst, J. M. Brown, A. F. Goncharov, and S. D. Jacobson, "Elasticity of (Mg,Fe)O through the spin transition of iron in the lower mantle" *Science* **319**, 451-453, (2008).
2. A. F. Goncharov, J. C. Crowhurst, and J. M. Zaug, "The elastic and vibrational properties of Co to 120 GPa," *Phys. Rev. Lett.* **92**, 115502 (2004).
3. J. C. Crowhurst, A. F. Goncharov, and J. M. Zaug, "Impulsive simulated light scattering from opaque materials at high pressure," *J. Phys Condensed Matter* **16**, S1137 (2003).
4. E. H. Abramson, J. M. Brown, L. J. Slutsky, and S. Wiryana, "Measuring speed of sound and thermal diffusivity in the diamond-anvil cell," *Int. J. Thermophys.* **22**, 405-414 (2001).
5. J. M. Zaug, E. Abramson, J. M. Brown, and L. J. Slutsky, "Sound velocities in olivine at Earth mantle pressures," *Science* **260**, 1487-1489, (1993).

Information Contacts: Joe Zaug (zaug1@llnl.gov) or Jonathan Crowhurst (Crowhurst1@llnl.gov)

Photoacoustic Light Scattering (PALS) – New Technique – 2010

Measuring Speeds of Sound –applied to virtually any fluid-state material



Alexander G. Bell first reported the photoacoustic effect in 1880 and 1881: Laser initiated photoacoustic light scattering (PALS) experiments began in the 1960's. Here we developed a new PALS geometry to generate and measures acoustic wave propagation from a wide class of transparent fluid materials. PALS offers technical advantages over conventional frequency-domain and time-domain techniques, (1) requires low average irradiance, between 1-3 orders of magnitude less than conventional laser-based sound velocity measurement techniques, which significantly reduces the potential for photochemical

reactions; (2) inherently low frequency measurements (approximately 0.5 - 5.0 GHz for fluids or 5 - 20 times lower than frequency-domain measurements when applied to high pressure fluids), which help to avoid or minimize liquid-state acoustic dispersion effects; (3) easily characterized acoustic dispersion, which can be conducted by symmetrically changing the incident and selected scattering wavevector of the probe beam, thus allowing determination of structural relaxation rates of polymers or glassy-like materials; (4) material index is not required to determine c ; and (5) permits the study of physically thin materials, e.g., <5 microns.

PALS publications to date:

1. J. M. Zaug, S. Bastea, J. C. Crowhurst, M. R. Armstrong, and N. Teslich Jr., "Photoacoustically Measured Speeds of Sound of Liquid HBO₂: Semi-Empirical Modeling of Boron-Containing Explosives," *J. Phys. Chem. Lett.* **1**, 2982 (2010)

https://www.llnl.gov/news/aroundthelab/2010/Oct/Explosives_Test.html

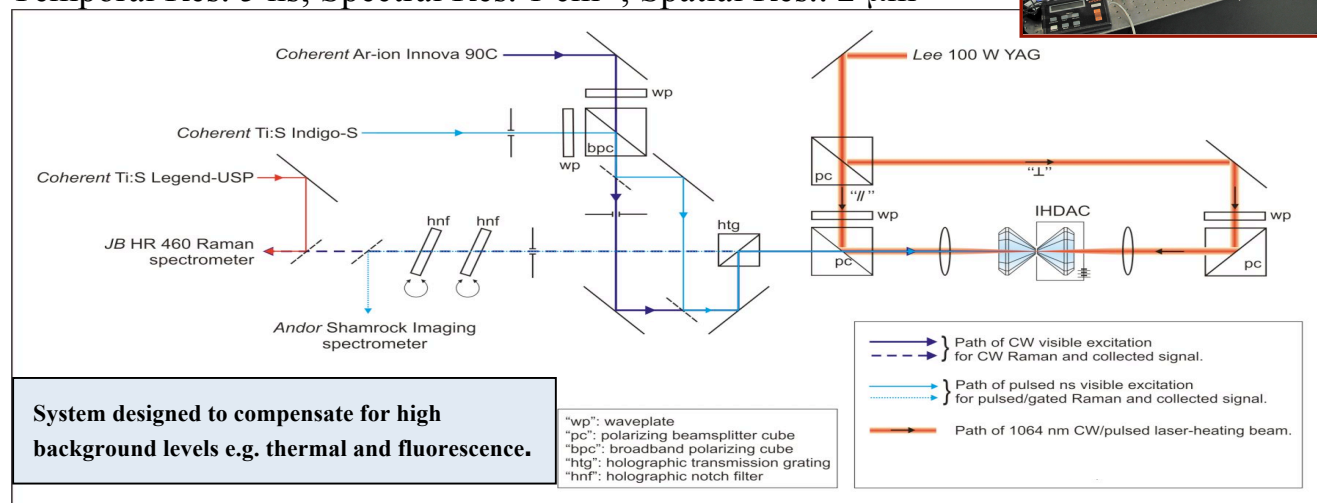
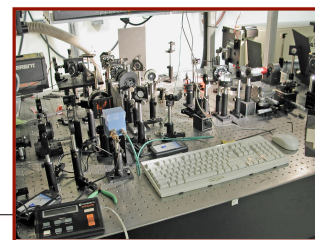
<http://rdmag.com/News/2010/10/Materials-Metals-Reinventing-Bells-photoacoustic-effect-for-explosives-study/>

2. J. M. Zaug, S. Bastea, J. C. Crowhurst, M. R. Armstrong, and N. E. Teslich Jr., "Photoacoustically measured speeds of sound and the equation of state of HBO₂: On understanding detonation with boron fuel," *Proceedings of the Fourteenth International Detonation Symposium*, Coeur d'Alene, ID, April, (2010). <http://www.intdetsymp.org/detsymp2010/GetFile.aspx?ID=34810&type=man>

CW and Pulse-Gated Raman Spectroscopy

Atomic Structures & Chemistry under Extreme Conditions

Temporal Res. 5 ns; Spectral Res. 1 cm^{-1} ; Spatial Res.: $2\text{ }\mu\text{m}$



Our CW and pulse-gated Raman instrument is designed to measure vibrational spectra under the most demanding circumstances where high signal background contributions are present. For example, the CW component has successfully enabled Raman studies where up to 2500 K thermal backgrounds are present (457 nm probe wavelength). Our pulse-gated Raman system extends the viable measurement range to $> 4000\text{ K}$. This system effectively eliminates sample fluorescence background contributions when electronic transition life times exceed 5 ns (minimum gate-width of our photo-tube). In addition, this system is elegantly designed to enable users to rapidly tune to multiple probe wavelengths available from our CW argon ion laser, e.g. 457 nm, 488 nm, 514 nm; our 632.8 nm HeNe source; and our 400-420 nm, 800-840 nm 20 ns pulse-width laser source (1 kHz maximum repetition rate). We also use a commercial Raman system to automate 3-D scans for enhanced spatial characterizations.

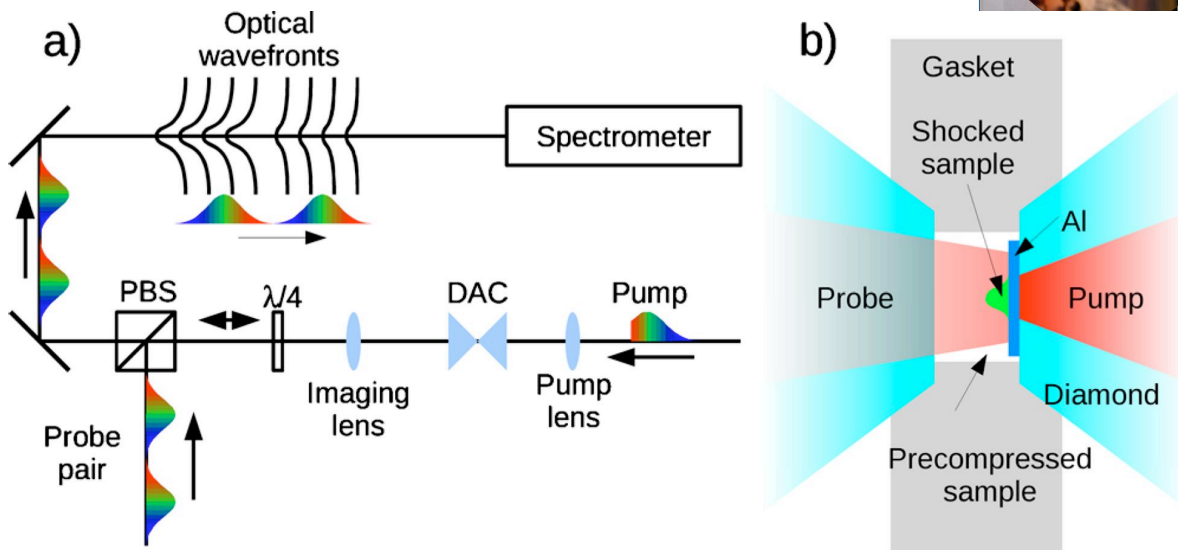
Selected Raman publications:

1. D. Aberg, P. Erhart, J. Crowhurst, J. M. Zaug, A. F. Goncharov, and B. Sadigh, "Pressure-induced phase transition in the electronic structure of palladium nitride," *Phys. Rev. B* **82**, 104116, (2010). [PRB Editors' Suggestion Paper \(September 2010\)](#)
2. A. F. Goncharov, J. C. Crowhurst, v. V. Struzkin et al., "Triple Point on the Melting Curve and Polymorphism of Nitrogen at High Pressure," *Phys. Rev. Lett.* **101**, 095502, (2008).
3. A. F. Goncharov, and J. C. Crowhurst, "Raman Spectroscopy of Hot Compressed Hydrogen and Nitrogen, Implications for the Intramolecular Potential," *Phys. Rev. Lett.* **96**, 055504, (2006).
4. J. C. Crowhurst, A. F. Goncharov, B. Sadigh et al. "Synthesis and Characterization of the Nitrides of Platinum and Iridium," *Science* **311**, 1275-1278, (2006).
5. A. F. Goncharov, and J. C. Crowhurst, "Pulsed Laser Raman Spectroscopy in the Laser Heated Diamond Anvil Cell," *Rev. Sci. Inst.* **76**, 063905 (2005).
6. A. F. Goncharov, N. Goldman, L. E. Fried, J. C. Crowhurst, I-Feng W. Kuo, C. J. Mundy, J. M. Zaug, "Dynamic Ionization of Water Under Extreme Conditions," *Phys. Rev. Lett.* **94**, 125508 (2005). See: Nature.com; <http://www.nature.com/news/2005/050321/full/050321-4.html>

Information Contacts: Jonathan Crowhurst (Crowhurst1@llnl.gov) or Joe Zaug (zaug1@llnl.gov)

Ultrafast Shock Interrogation (USI)

Reflectivity, Shock & Particle Speeds, Transient State Studies, Time of Flight Acoustics, Nonthermal Phase Transitions, THz Temporal Res. 0.2 ps; Window 272 ps; Spatial Res. 0.5 μm



Our ultrafast shock interrogation diagnostics enable characterization of phenomena that occur on the picosecond or greater timescales. We use USI to investigate shock induced phenomena in metals and transparent media. This method is very similar to VISAR, but USI works on an ultrafast time scale: It provides time dependent phase measurements between a probe and reference reflected from a shocked sample as shown in the above Figure b). Chirp pulse amplified pump pulses are tightly focused onto a 1-2 micron thick aluminum, (Al) film, which coats one face of a substrate window or sample. In the case of precompression, up to 100 GPa may be obtained prior to a USI shot. Pump-induced thermal expansion, over the deposition depth of an Al film, launches a shock wave that travels through the film and enters a sample or pressure medium. We determine (from the time dependent phase of a reflected probe) shock velocity, particle velocity, and the index of refraction change at a shock front. The USI capability currently generates up to 50 GPa shock waves in a DAC at precompressions of >50 GPa. We also conduct time-of-flight acoustic wave measurements at precompressions up to 50 GPa. We continue to explore strain induced terahertz radiation signatures.

USI publications to date:

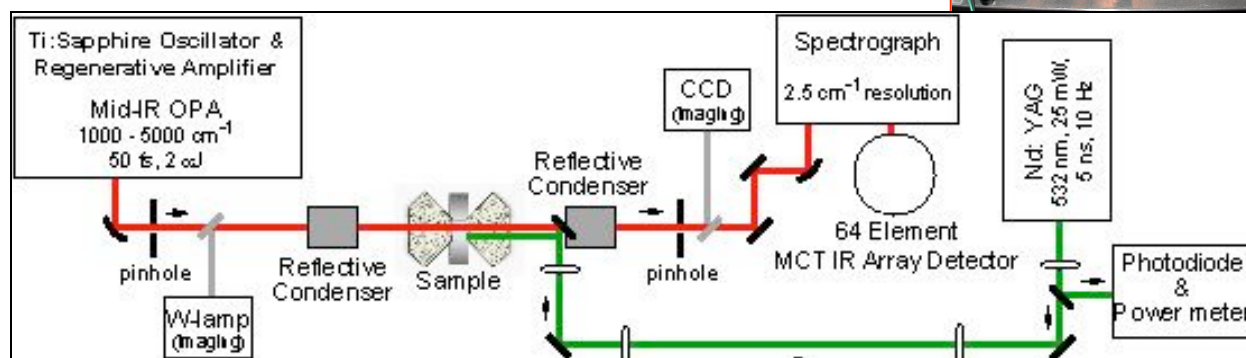
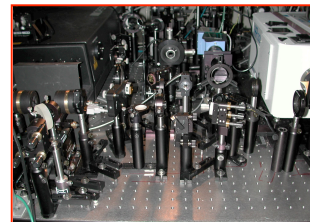
1. M. R. Armstrong, J. C. Crowhurst, S. Bastea and J. M. Zaug, "Ultrafast observation of shocked states in a precompressed material," *J. Appl. Phys.* **108**, 023511, (2010).
https://publicaffairs.llnl.gov/news/news_releases/2010/NR-10-07-01.html
2. M. R. Armstrong, J. C. Crowhurst, E. J. Reed, and J. M. Zaug, "Ultrafast high strain rate acoustic wave measurements at high pressure in a diamond anvil cell," *App. Phys. Lett.* **92**, 101930, (2008).
3. E. J. Reed, M. R. Armstrong, K. Y. Kim et al. "Atomic-scale time and space resolution of terahertz frequency acoustic waves," *Phys. Rev. Lett.* **101**, 014302, (2008).
4. E. J. Reed, M. R. Armstrong, K. Y. Kim, et al. "Terahertz radiation from shocked materials," *Mat. Today* **10**, 44, (2007).

Contacts: Mike Armstrong armstrong30@llnl.gov , crowhurst1@llnl.gov , zaug1@llnl.gov

Time Resolved Infrared Absorption (TRIR)

Transient Chemistry, Transient Atomic Structures

Temporal Resolution: 50 fs; Spatial Resolution 20 μm



Our TRIR instrument monitors an initiated chemical and/or physical sequence with a time resolution of >50 femto seconds. This system is configured for transmission studies, however TRIR may also be conducted in reflection mode. At present, a 7 ns pulse width laser-drive initiation system or a 100 W CW laser heating source can be focused onto encapsulated samples. More recently we have initiated reactions using a custom-made pulsed plasma drive system. In principal it is possible to use a portion of the regenerative amplifier output (50 fs pulse width at 800 nm or 400 nm) to either photo initiate or photo-mechanically initiate a structural and/or chemical process. Our TRIR system operates in single-shot mode or repetitively at maximum rate of 1.0 kHz. The accessible spectral range is 2-10 microns ($5000 - 1000 \text{ cm}^{-1}$). The one-inch working distance of our Cassegrain optics enables the use of exceptionally long and/or thermally heated sample holders. This experimental system is the first-ever to generate and monitor ns transient chemistry within a high-pressure diamond-anvil cell.

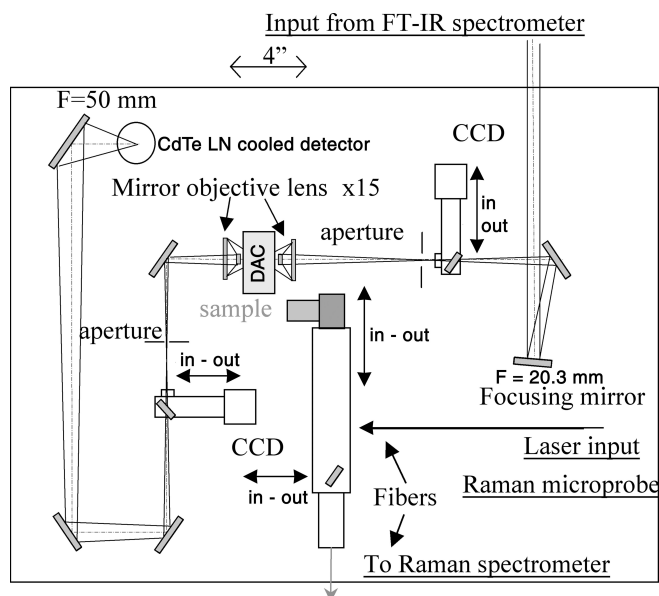
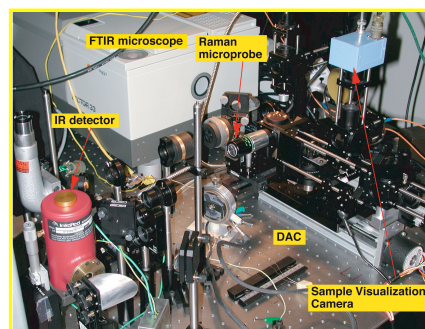
TRIR group publications to date:

1. E. A. Glascoe, J. M. Zaug et al., "Nanosecond time resolved infrared studies of photoinduced decomposition of TATB at ambient and elevated pressures," *J. Chem. Phys. A* **113**, 5503, (2009).
2. E. A. Glascoe, K. R. Sawyer et al., "The Influence of the metal spin state in the iron-catalyzed alkene isomerization reaction studied with ultrafast infrared absorption," *J. Phys. Chem. C* **111**, 8789, (2007).
3. J. E. Shanoski, E. A. Glascoe, and C. B. Harris, "Ligand rearrangement reactions of $\text{Cr}(\text{CO})_6$ in alcohol solutions" Experiment and Theory," *J. Chem. Phys. B.* **110**, 996, (2006).
4. M. F. King, J. F. Cahoon, E. A. Glascoe et al., "The role of odd-electron intermediates and in-cage electron transfer in ultrafast photochemical disproportionation reactions in Lewis bases," *J. Am. Chem. Soc.* **126**, 11414, (2004).

Information Contacts: E. (Libby) Glascoe (Glascoe2@llnl.gov) or Joe Zaug (zaug1@llnl.gov)

Fourier Transform Infrared Absorption

Chemical Kinetics; Phase Stability; Atomic Structures
Spatial Resolution: 20 μm



Our FTIR instrument monitors slow (seconds to hours) transient chemical and/or physical transitions. This system is configured for transmission studies; however, FTIR may also be conducted in reflection mode. The accessible spectral range is 1-20 microns ($10000 - 500 \text{ cm}^{-1}$). The one-inch working distance of the Cassegrain optics enables the use of heated sample and/or high-pressure sample holders. A μ -Raman and/or fluorescence system is embedded within our FTIR system to enable additional near-

simultaneous physical and/or chemical characterizations including pressure determination using optical-type manometers. UV/Vis absorption measurements are also conducted using this system.

Selected FTIR publications and manuscripts:

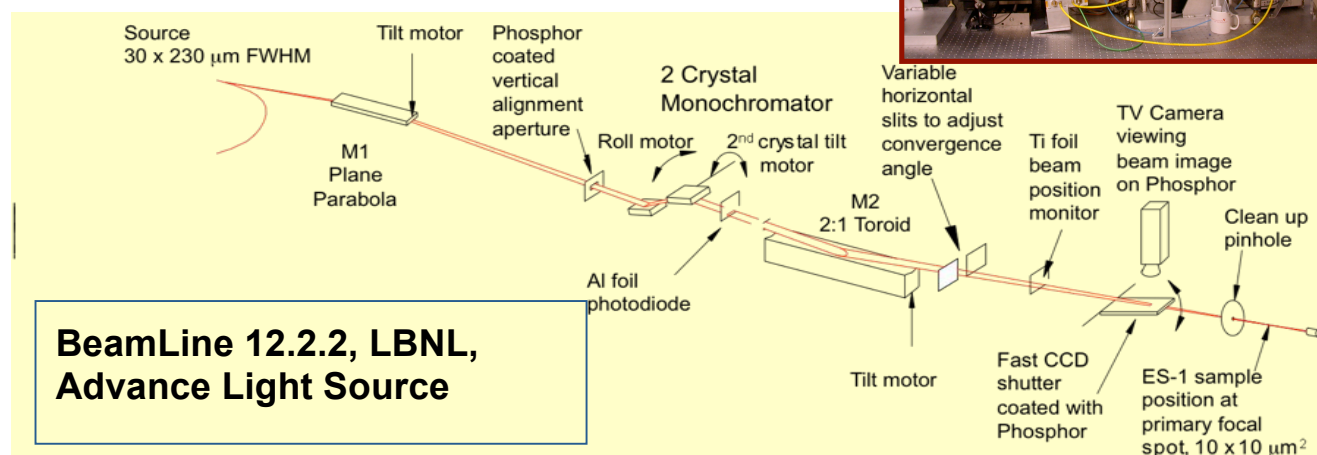
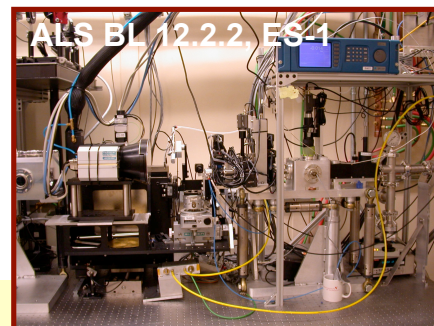
1. E. A. Glascoe, J. M. Zaug A. K. Burnham, "Pressure dependent decomposition kinetics of the energetic material HMX," *J. Phys. Chem. A* **113**, 13548, (2009).
2. W. Montgomery, J. C. Crowhurst, J. M. Zaug, and R. Jeanloz, "The chemistry of cyanuric acid ($\text{H}_3\text{C}_3\text{N}_3\text{O}_3$) under high pressure and high temperature," *J. Phys. Chem. B* **112**, 2644, (2008).
3. A. F. Goncharov, M. R. Manaa, J. M. Zaug, R. H. Gee, L. E. Fried, and W. B. Montgomery, "Polymerization of formic acid under pressure," *Phys. Rev. Lett.* **94**, 065505, (2005).
Highlighted in the journal Nature, (March 2005)
4. W. Montgomery, J. M. Zaug, M. H. Howard, A. F. Goncharov, J. C. Crowhurst, and R. Jeanloz, "The melting curve and high pressure chemistry of formic acid to 8 GPa and 600 K," *J. Phys. Chem. B* **109**, 19443 (2005).

Information Contacts: Joe Zaug (zaug1@llnl.gov) or E. (Libby) Glascoe (Glascoe2@llnl.gov)

Synchrotron X-Ray Diffraction

Amorphous & Crystalline Material, Structural Kinetics
Atomic & Void Structures, EOS, Bulk Moduli

High brilliance, spatial resolution = 10 μm .



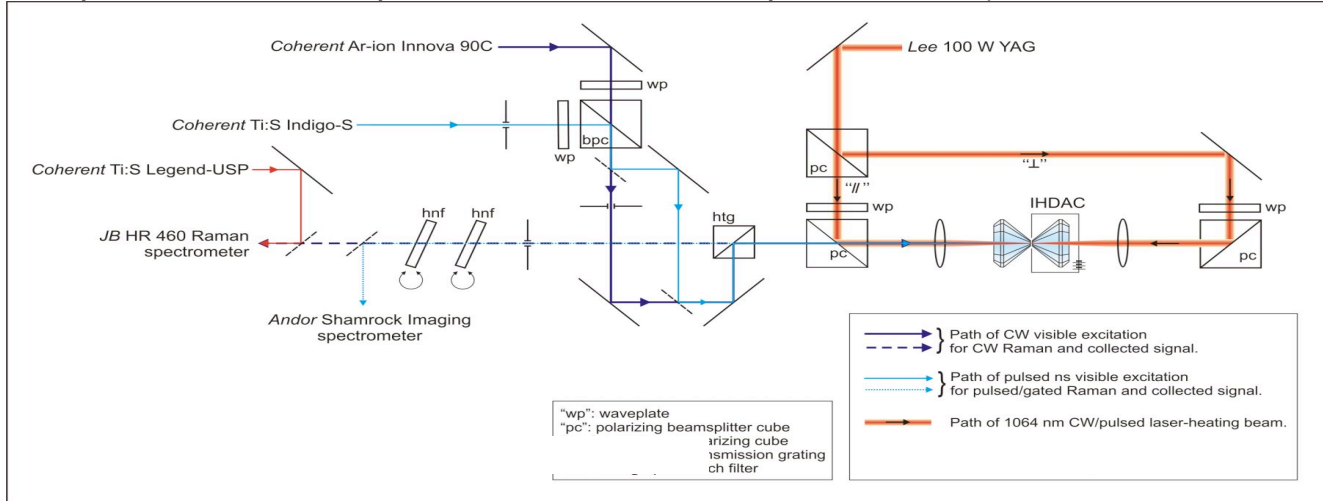
Our group utilizes 3rd generation synchrotron radiation sources to characterize atomic structures of crystalline and disordered materials subjected to high pressure and/or temperature conditions. The facilities we use include the, APS (6 GeV, ANL), ALS (2 GeV, LBNL), SSRL (3 GeV, Stanford), ESRF (6 GeV, Grenoble, France), NSLS (2.8 GeV, BNL), and SPRING-8 (8 GeV, Hyogo, Japan). Each source provides dedicated high-pressure materials science end stations with added capabilities that include CW laser heating and optical characterization diagnostics such as Raman and photoluminescence spectroscopy. In addition, we use neutron sources such as LANCE at LANL, which are key toward determining the atomic positions of light elements such as hydrogen.

Selected XRD publications:

1. S. M. Clark, and J. M. Zaug, "The compressibility of cubic white, orthorhombic black and rhombohedral black phosphorus," *Phys. Rev. B* –Accepted for publication August (2010).
2. D. Aberg, P. Erhart, J. Crowhurst, J. M. Zaug, A. F. Goncharov, and B. Sadigh, "Pressure-induced phase transition in the electronic structure of palladium nitride," *Phys. Rev. B* **82**, 104116 (2010).
[PRB Editors' Suggestion Paper \(September 2010\)](#)
3. C. D. Grant, J. C. Crowhurst, Arsenlis T., Bringa E. M. et al., "X-ray diffraction of electrodeposited nanocrystalline nickel under pressure," *J. Appl. Phys.* **105**, 084311, (2009).
4. J. M. Zaug, A. K. Soper, and S. M. Clark, "Pressure-dependent structures of amorphous red phosphorus and the origin of the first sharp diffraction peaks," *Nature Materials* **7**, 890, (2008). J. C. Crowhurst, A. F. Goncharov, B. Sadigh et al. "Synthesis and characterization of the nitrides of platinum and iridium," *Science* **311**, 1275, (2006).
5. A. F. Goncharov, M. R. Manaa, J. M. Zaug, R. H. Gee, L. E. Fried, and W. B. Montgomery, "Polymerization of formic acid under pressure," *Phys. Rev. Lett.* **94** 065505, (2005).
[Highlighted in the journal Nature \(March 2005\)](#)

Information Contacts: Joe Zaug (zaug1@llnl.gov) or Jonathan Crowhurst (crowhurst1@llnl.gov)

Temporal Res. 5 ns; Spectral Res. 0.01 nm, Spatial Res. 2 μm



Selected Photoluminescence publications:

1. C. D. Grant, J. C. Crowhurst, S. Hamel et al., "Anomalous photoluminescence in CdSe quantum-dot solids at high static pressure in a diamond anvil cell," *Small* **4**, 788, (2008).
2. F. Wu, J. M. Zaug, C. E. Young, and J. Z. Zhang, "Pressure-induced phase transition in Thiol-capped CdTe Nanoparticles," *J. Nanosci. Nanotech.* **8**, 6528, (2008).
3. A. F. Goncharov, J. M. Zaug, and J. C. Crowhurst, "Optical calibration of pressure sensors for high pressures and temperatures," *J. Appl. Phys.* **97**, 094917 (2005).
4. J. C. Crowhurst, I. M. Darnell, A. F. Goncharov, D. H. Lassila, and J. M. Zaug, "Determination of the coefficient of friction between metal and diamond under high hydrostatic pressure," *Appl. Phys. Lett.* **85**, 5188 (2004).

Information Contacts: Chris Grant (grant29@llnl.gov) or (crowhurst1@llnl.gov), (zaug1@llnl.gov)

List of Collaborators

More than 60 collaborators over the last 48 months

List of Collaborators and Co-Authors and Current Affiliations (last 48 months):

D. Aberg	Lawrence Livermore National Laboratory, Livermore, CA
M. Ahart	Carnegie Institute in Washington D.C.
A. P. Alivisatos	Lawrence Berkeley National Laboratory, Berkeley, CA
P. Allen	Lawrence Livermore National Laboratory, Livermore, CA
M. R. Armstrong	Lawrence Livermore National Laboratory, Livermore, CA
B. Baer	Lawrence Livermore National Laboratory, Livermore, CA
J. D. Bass	University of Illinois, Urbana-Champaign, IL
S. Bastea	Lawrence Livermore National Laboratory, Livermore, CA
T. F. Baumann	Lawrence Livermore National Laboratory, Livermore, CA
P. Beck	Carnegie Institute in Washington D.C.
J. M. Brown	University of Washington, Seattle, WA
A. K. Burnham	American Shale Oil LLC, Rifle, CO
O. Cervantes	Lawrence Livermore National Laboratory, Livermore, CA W.
Carr	Lawrence Livermore National Laboratory, Livermore, CA S.
M. Clark	Lawrence Berkeley National Laboratory, Berkeley, CA
J. C. Crowhurst	Lawrence Livermore National Laboratory, Livermore, CA D.
A. Dixon	University of Alabama, Tuscaloosa, AL
P. Erhart	Lawrence Livermore National Laboratory, Livermore, CA
Y. Fei	The City University of New York, New York, NY
M. F. Foltz	Lawrence Livermore National Laboratory, Livermore, CA
L. E. Fried	Lawrence Livermore National Laboratory, Livermore, CA
A. E. Gash	Lawrence Livermore National Laboratory, Livermore, CA
J. K. Gibson	Lawrence Berkeley National Laboratory, Berkeley, CA
N. Goldman	Lawrence Livermore National Laboratory, Livermore, CA
E. A. Glascoe	Lawrence Livermore National Laboratory, Livermore, CA
A. E. Gleason	University of California, Berkeley, CA
A. F. Goncharov	Carnegie Institute in Washington D.C.
C. D. Grant	Lawrence Livermore National Laboratory, Livermore, CA
E. K. Hart	Stanford University, Palo Alto, CA
W. M. Howard	Lawrence Livermore National Laboratory, Livermore, CA
S. Jacobsen	Northwestern University, Evanston, IL
R. Jeanloz	University of California, Berkeley, CA
J. Kenneally	Lawrence Livermore National Laboratory, Livermore, CA
B. Khare	NASA AMES laboratory, Mnt. View, CA
I.-F. Kuo	Lawrence Livermore National Laboratory, Livermore, CA
M. Kunz	Lawrence Livermore National Laboratory, Livermore, CA

D. Lakshtanov	University of Illinois, Urbana-Champaign, IL
J.-F. Lin	University of Texas, Austin, TX
J. L. Maienschein	Lawrence Livermore National Laboratory, Livermore, CA
C. McKay	NASA AMES Laboratory, Mnt. View, CA
M. R. Manaa	Lawrence Livermore National Laboratory, Livermore, CA Y.
Meng	Argonne National Laboratory, Argonne, IL
R. Miles	Lawrence Livermore National Laboratory, Livermore, CA
B. Militzer	University of California, Berkeley, CA
K. J. Moody	Lawrence Livermore National Laboratory, Livermore, CA
T. J. Norman	Stanford Research Institute, Menlo Pk. CA
S. Peiris	Defense Threat Reduction Agency, Fort Belvoir, MD
V. B. Prakapenka	Argonne National Laboratory, Argonne, IL
Peter J. Pauzauskie	University of Washington, Seattle, WA
J. E. Reaugh	Lawrence Livermore National Laboratory, Livermore, CA
E. J. Reed	Stanford University, Palo Alto, CA
T. Rose	Lawrence Livermore National Laboratory, Livermore, CA
B. Sadigh	Lawrence Livermore National Laboratory, Livermore, CA
J. H. Satcher	Lawrence Livermore National Laboratory, Livermore, CA
W. Seikhause	Lawrence Livermore National Laboratory, Livermore, CA
D. Shaughnessy	Lawrence Livermore National Laboratory, Livermore, CA
S. Sinogeikin	University of Illinois, Urbana-Champaign, IL
A. K. Soper	Rutherford Appleton Laboratory, UK
H. K. Springer	Lawrence Livermore National Laboratory, Livermore, CA
Nick Teslich Jr.	Lawrence Livermore National Laboratory, Livermore, CA
S. N. Tkachev	University of Hawaii, Honolulu, Hawaii
B. Watkins	Lawrence Livermore National Laboratory, Livermore, CA
W. Wilson	Defense Threat Reduction Agency, Fort Belvoir, MD
M. A. Worsley	Lawrence Livermore National Laboratory, Livermore, CA
F. Wu	University of California, Berkeley, CA
J. Z. Zhang	University of California, Santa Cruz, CA

Extreme Chemistry Experimental Group Projects

Selected studies from our R&D portfolio

Science in the National and Global Interest

October 2010

- **STOCKPILE STEWARDSHIP:** We collaborate with LLNL experts in computational chemistry to predict the performance of energetic materials formulations. Our speeds of sound results facilitate development of materials equations-of-state (EOS), which are required to semi-empirically predict e.g., using the CHEETAH thermochemical code, thermodynamic states at detonation, deflagration, and combustion conditions. We also determine shock Hugoniot and, when combined with diamond-anvil cell technology, off-Hugoniot state properties of a wide range of materials using our ultrafast shock interrogation technique (USI). Moreover, USI provides a window into physico-chemical transitions that occur on the picosecond timescale. Shock-synthesis of new materials is also explored. By integrating our experimental and computational capabilities and expertise, we systematically advance science-based stockpile stewardship agendas, provide guidance to large-scale energetic materials formulation tests, and elucidate thermochemical processes of direct relevance to global security.
- **NUCLEAR FORENSICS AND ENERGY SCIENCES:** We provide experimental evidence to guide nuclear materials forensics attribution efforts. Currently we are characterizing the high-temperature chemistry of UO_2 reactant when in the presence of common gas and fluid specie environments. Here we utilize our custom-made vibrational spectroscopy systems and key ex situ characterization tools, to characterize uranium chemistry that is likely to occur in engineer-controlled environments or under explosive conditions. Ultimately this work provides fundamental data required to optimize the safety and design performance of nuclear power plant systems. In addition, our results provide insight to the nuclear forensics/attribution community: It is important to develop a priori knowledge of the chemical partitioning of UO_2 , occurring at temperatures representative of critical or subcritical detonations.
- **CHEMICAL ENERGY SYSTEMS:** The ability to synthesize and characterize novel compounds that store significant amounts of chemical energy -with on-demand energy release- is an increasingly important scientific and engineering issue. The motivation for our research stems in part by ongoing reductions of our planetary fossil fuel energy reserves concomitant with production of greenhouse gas production. As part of a search for "green" chemical energy systems, we synthesize polynitrogen compounds at high temperatures and modestly low pressures. In collaboration with LLNL computational chemistry experts, our experimental results are used to guide the characterization of material structures, physico-chemical stability fields, and potential energy and release performance.

- **CHARACTERIZATION of IMPROVISED EXPLOSIVES:** The threat of improvised explosives (IEs) continues to challenge our global and national security. It is important to recognize to what extent detonability can be achieved with IEs. Here we study IEs to quantify physical and chemical parameters and elucidate thermodynamic states that make the difference between deflagration (low damage yield) and detonation (high damage yield). Our semi-empirical assessments of detonability, grounded in speeds of sound and molecular phase stability measurements, provide rapid and accurate results that may otherwise be difficult to obtain by large-scale trial-and-error tests. Moreover, our approach is potentially applicable to myriad classes of IE formulations e.g., liquids, solids, and mixtures.

Applied Research

- **HYDROGEN and DEUTERIUM:** We have been awarded a LLNL Laboratory directed research and development (LDRD) grant to shock precompressed hydrogen or deuterium. Samples are first brought up to many 10's of GPa using conventional diamond-anvil cell technology, and then they are mechanically shocked using our USI method. The aim is to characterize these materials at high-density shock states, which are not accessible by any known experimental methods. Our results will guide the development of fundamental and theoretical knowledge of simple molecular interactions –how they proceed at conditions relevant to myriad of energetically dynamic processes. Phase stability curves will be delineated. There is the possibility of transitioning these materials to the metallic state.
- **THOLIN CHEMISTRY and TITAN'S METHANE MYSTERY:** In collaboration with scientists at NASA's Ames Research Center and LLNL computational chemistry experts, we are attempting to resolve a long-standing issue regarding the presence of methane (5 % by volume) in Titan's atmosphere. The lifetime of CH₄ on Titan is estimated to be 10 – 100 million years. Numerous scientific studies have effectively ruled out many possible production mechanisms; hence, one is lead to believe that methane is produced below the surface. Using our custom-designed micro-FTIR instrumentation and, mass spectrometry capabilities and expertise at NASA, we will characterize pressure and/or temperature induced products formation from Titan-Tholin starting material.
- **TERAHERTZ RADIATION SOURCES:** We are funded by DARPA to investigate methods to amplify and modulate THz acoustic waves in materials, ultimately in the interest of developing more versatile THz radiation sources. Previously, we developed a method to coherently generate THz radiation from a THz acoustic wave (Armstrong et al., Nature Physics 2009); however, this method requires ultrafast laser system to generate THz acoustic waves. Here we are exploring different approaches to generate and launch THz acoustic waves where large ultrafast systems are not required. A portable THz generator would also facilitate spatial imaging of materials with nanometer length scale resolution.